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Dynamic behavior of azobenzene polyester used for holographic data storage

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ABSTRACT

Dynamic behavior of thin photoaddressable polyester films was studied. The saturation process due to Fourier holographic recording was investigated. Model experiments show an optimal intensity ratio of the object and reference beams, where the highest efficiency occurs. This ratio is inversely proportional to the reference intensity. The material has a significantly higher sensitivity at 407 nm than at 532 nm. For 1 μm thick sample an M# of 0.25 was measurable.

INTRODUCTION

Photoaddressable polymers [1] are candidate materials for holographic data storage. Such polymers are being developed at Risø National Laboratory in Denmark [2,3]. High storage density can be achieved by recording Fourier hologram of the binary object. Similar method is used in many applications [4,5]. The presence of high intensity peak at the Fourier plane saturates the recording material therefore a phase mask or shifting from the Fourier plane has to be used [5]. The second solution causes a significant decrease in the storage density. The recording system can be optimized by determining dynamic behavior of the recording material. Experiments with high intensity ratio plane waves were carried out for recording and demonstrating the saturation effect due to recording the Fourier transformed data page. The first section of the paper deals with these experiments.

The storage capacity can be further enhanced by reducing the writing wavelength and by multiplexing. The sensitivity factor of the material has been determined at two different wavelengths (532 and 407 nm). Multiplexing feature of the storage material is usually represented by the M# [6], a property dependent on both writing and erasure dynamics. We carried out several experiments in order to determine it. In the second and third section, we describe the investigation of the wavelength dependent sensitivity and multiplexing ability of the applied material.

SATURATION OF THE POLYMER

In our holographic memory system polarization holography is used [7] and the Fourier transformed data page is recorded by using orthogonal circularly polarized object and reference beams in an appropriate 8f arrangement [4]. A suitable model setup (see figure 1) was built to study the behavior of the material under extreme dynamic circumstances like recording Fourier holograms. Anisotropy gratings were recorded in the storage material using beams with

orthogonal circular polarizations and high intensity ratios. An intensity ratio of 500 has been achieved, which easily occurs when Fourier holograms are recorded.

There were two limiting factors in adjusting proper intensity ratio beams in the experiment. The maximum laser power (frequency doubled Nd:YAG, $\lambda = 532$ nm, $P_{output} = 50$ mW) and the detectability of the recorded holograms determined the upper and lower limits, respectively. These requirements provided the maximum and minimum applied intensities of the recording. We decreased the hologram size by using a lens (f = 140mm) to reach the maximum intensity. Since the hologram radius was small (≤ 0.27 mm) compared to the focal length of the lens, instead of plane waves, beams with nearly planar wavefronts and with negligible radius of curvature were used.
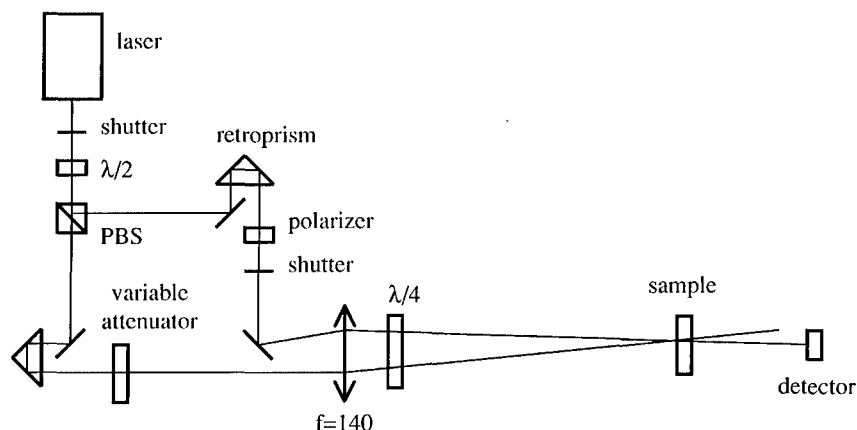

Figure 1. Experimental setup for recording polarization holograms with high intensity ratio beams, i.e. $I_{obj}/I_{ref} \leq 500$.

For the compensation of low coherence length laser (~1-2 mm) two retroprisms were used. The reference and object arms were set for optimum interference condition when maximum modulation of the interference fringes occurs, i.e. when the diffracted intensity has its maximum value.

It is well known that the process of writing polarization holograms in azobenzene polymers is often followed by creating surface grating [3,8] besides anisotropy grating in the bulk material. Since the creation of surface grating is slow [3] at the applied intensities, a suitable short exposure time ($t_{exp} = 0.5$ sec) was used. This exposure time satisfied also the requirement of detectability providing measurable diffracted power. Every hologram was made on the same place of the sample in order to get rid of sample inhomogeneity.

For readout we used the arm equipped with the attenuator in order to maximize the diffracted power. The power of the readout beam was the same for every experiment. Since the readout causes erasure of the hologram, the maximum value of the diffraction efficiency was measured by a detector connected to the computer by means of a fast digitizing card.

The recording material was an amorphous side-chain azobenzene polyester (E1aP) spincoated onto a glass sample. E1aP is prepared through melt transesterification of 3-[4-((4-

cyanophenyl)azo)-phenoxy]-1,2-propanediol and diphenyl phthalate. The thickness of the layer was 2 μm covered with a hard protective SiO_2 layer.

Two sets of measurement were carried out using two different hologram sizes (applied Gaussian beam radii were $w = 0.27$ mm and 0.11 mm). Figure 2 shows measured diffraction efficiency as a function of the intensity ratio of the object and reference beams for the larger hologram size (parameter: reference intensity). There is an optimum intensity ratio for a given reference intensity where the highest diffraction efficiency occurs. This optimum intensity ratio is inversely proportional to the reference intensity.
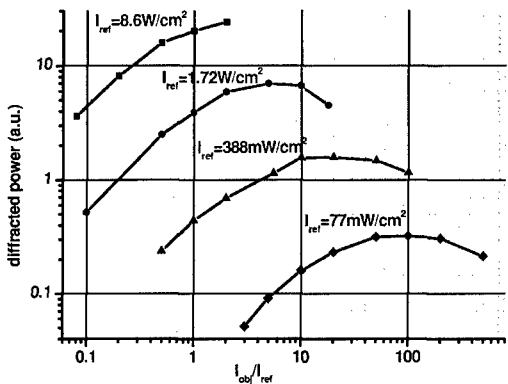

Figure 2. Measured diffracted power as a function of the intensity ratio of the object and reference beams at different reference intensities, radius of beams is 0.27 mm.
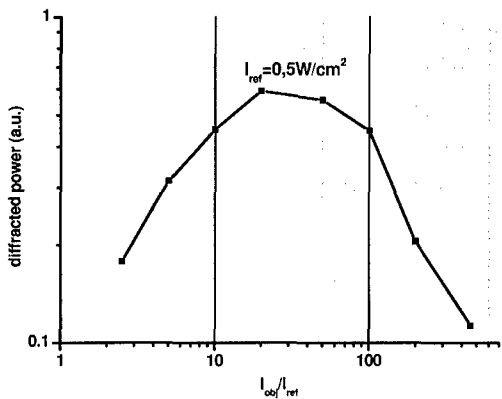

Figure 3. Diffracted power as a function of the intensity ratio of object and reference beams with beam radius of 0.11 mm.

Figure 3 shows results for the smaller hologram size (w = 0.11 mm), here the decreasing part of the diffracted power can is also be seen. The strong decay in the diffracted power for high intensity ratios is due to the saturation of the material.

BLUE SENSITIVITY

There is a strong demand in the optical data storage technology to apply the shortest available wavelength for recording in order to enhance the storage density. In our holographic memory system a frequency doubled Nd:YAG laser was used so far. The appearance of violet laser diode on the photonics market has opened the opportunity of using compact blue laser source for recording. Before the modification of the whole system, determination of the blue response of our polymer has particular importance.

Appropriate polarization holographic setups (similar to the setup in figure 1) were built for comparing blue and green responses of two different polymers (E1aP and E1aP/12) when using transmissive and reflective samples. E1aP12 is a three component polyester that contains both aromatic and aliphatic residues in the main chain.

A Kr^+ laser operating at 407 nm and a frequency doubled Nd:YAG laser at 532 nm were used for writing and we used a He-Ne laser at 633 nm for the readout in order to measure the diffraction efficiency in real time. The polymer has no absorption at this wavelength, so the readout in this region does not cause any erasure.

Time dependent exposures were performed at different intensities in order to compare the sensitivity of the material at different wavelength. Our goal was to determine the thickness independent sensitivity value S defined as usual

$$S = \frac{\sqrt{\eta}}{I \cdot t} \qquad [cm^2/J] \qquad (1)$$

where η is the measured diffraction efficiency at the writing intensity of I when using an exposure time of t. This value is often used in [9] in order to compare different holographic materials. Table I shows calculated sensitivity values of the azobenzene polymer samples when $\eta = 0.5\%$ was taken for comparison.

Table I. Sensitivity values of different transmissive and reflective polymer samples at the two wavelengths (d is the layer thickness).

Sample type and thickness / wavelength $[cm^2/J]$		407 nm	532 nm
Transmissive	E1aP, d = 0.9 μm	0.182	0.016
	E1aP/12, d = 1.4 μm	0.245	0.034
Reflective	E1aP, d = 1 μm	0.142	0.047
	E1aP/12, d = 1 μm	0.230	0.050

Table I shows increased sensitivity of the azobenzene polymers at 407 nm. The ratio of the sensitivity values at the two wavelengths is between 3 and 11 depending on the polymer type and thickness. The increased sensitivity has a beneficial consequence to the writing speed at 407 nm.

MULTIPLEXING ABILITY: M#

A relevant feature of the holographic recording material is its multiplexing ability represented by the M#. This number characterizes the material and the applied experimental arrangement. There are two ways to determine it [6]. The first method is to make a large number of multiplexing (M) with a proper exposure schedule that results in equal hologram efficiencies. In this case

$$\eta = (M\#/M)^2 \tag{2}$$

The other method is based on the assumption that each hologram evolves during recording as $A_0[1-\exp(-t/\tau_{rec})]$. This can be approximated at the initial time with A_0*t/τ_{rec} and the hologram decays during erasure as $\exp(-t/\tau_{erase})$. A_0 is the grating strength of the hologram, τ_{rec} and τ_{erase} are the time constants of recording and erasing, respectively. The ratio (A_0/τ_{rec}) and the time constant (τ_{erase}) can be determined by fitting appropriate parts of the experimentally measured $\sqrt{\eta}$ during recording and erasing, respectively. It is important that the writing and erasing intensity must be the same considering the intensity circumstances of multiplexing. The exact expression of the M# number is as follows:

$$M\# = (A_0/\tau_{rec})\,\tau_{erase} \tag{3}$$

We used the second method for the determination of the M#. Several exposures were made and each of them was followed by erasure. The intensity was equally divided between the writing beams, so the erasing intensity was the half of the writing intensity. In order to be compatible with the theory, we made two holograms on each sample with 1 W/cm^2 and 2 W/cm^2 intensity. When calculating the M#, the evolving (A_0/τ_{rec}) factor was calculated from the measurement at I = 1 W/cm^2 writing intensity and the decay (τ_{erase}) factor calculated from the measurement at 2 W/cm^2 writing intensity. The experiments were carried out at both 532 nm and 407 nm either. Table II shows these values and the calculated M# factors.

Table II. M# for different polymer samples.

Transmissive samples		407 nm			532 nm		
		A_0/τ_{rec}	τ_{erase}	M#	A_0/τ_{rec}	τ_{erase}	M#
E1aP	I = 1 W/cm^2	0.22	1.6	0.202	0.025	8.04	0.126
d = 0.9 μm	I = 2 W/cm^2	0.42	0.92		0.032	5.04	
E1aP/12	I = 1 W/cm^2	0.22	2.25	0.279	0.038	10.68	0.319
d = 1.4 μm	I = 2 W/cm^2	0.43	1.27		0.049	8.4	
Reflective samples		407 nm			532 nm		
		A_0/τ_{rec}	τ_{erase}	M#	A_0/τ_{rec}	τ_{erase}	M#
E1aP	I = 1 W/cm^2	0.12	4.22	0.214	0.068	3.78	0.205
d = 1 μm	I = 2 W/cm^2	0.28	1.78		0.090	3.02	
E1aP/12	I = 1 W/cm^2	0.22	2.56	0.264	0.055	10.04	0.265
d = 1 μm	I = 2 W/cm^2	0.35	1.2		0.095	4.82	

M numbers in table II are in the range of 0.25 and there is no difference for the two different wavelengths. However, these values are not small considering the thickness of the layer. Similar values were reported for photoaddressable and photorefractive polymers by [10], but they are still low compared to the M# of thick photorefractive materials [9].

CONCLUSIONS

High storage density can be achieved by recording a Fourier hologram of a binary object. However, the high intensity peak in the Fourier plane saturates the recording material. Model experiments were performed to study saturation behavior of azobenzene polyester material. Results show that the material provides maximum diffraction efficiency when optimum intensity ratio is used for the reference and object beams. The diffraction efficiency strongly decreases when high intensity ratio beams are applied.

Sensitivity factor and multiplexing ability of the materials were studied at two different wavelengths. Results show an increased sensitivity of the materials at 407 nm. For the 1 μm thick samples, the calculated M# values are about 0.25 for the two wavelengths.

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